

Claims

[c1] CLAIMS:

1. A curable composition, comprising:

- (a) an epoxy resin and curing agent therefor, wherein said epoxy resin is essentially free of bromine atoms;
- (b) a flame retardant additive essentially free of phenolic groups and of epoxy groups, wherein said flame retardant is a condensation product of (i) a brominated phenol or a mixture of brominated phenols with (ii) a cyanuric halide;
- (c) a thermoplastic resin; and
- (d) a cyanate ester.

[c2] 2. The curable composition of claim 1, wherein said flame retardant additive has a bromine content greater than 20%.

[c3] 3. The curable composition of claim 1, wherein said flame retardant additive is 1,3,5-tris(2,4,6-tribromophenoxy)triazine.

[c4] 4. The curable composition of claim 1, wherein said flame retardant additive is 2,2'-[(1-methylethylidene)bis[(2,6-dibromo-4,1-phenylene)oxy]]bis[4,6-bis[(2,4,6-tribromophenyl)oxy]-1,3,5-triazine].

[c5] 5. The curable composition of claim 1, wherein said flame retardant additive is soluble in toluene at a concentration of greater than 15 g/100ml of toluene at a temperature of 50 ° C.

[c6] 6. The curable composition of claim 1, wherein said epoxy resin is a glycidyl ether resin or a mixture of glycidyl ether resins containing, on average, greater than 2 epoxy groups per molecule.

[c7] 7. The curable composition of claim 1, wherein said epoxy resin is a mixture of:
(a1) an epoxy resin containing on average less than or equal to 2 glycidyl groups per molecule; and
(a2) an epoxy resin containing greater than 2 glycidyl groups per molecule.

[c8] 8. The curable composition of claim 1, wherein said thermoplastic resin has a

Tg greater than 120 ° C.

- [c9] 9. The curable composition of claim 1, wherein said thermoplastic resin has a dissipation factor of less than 0.010 measured at 1 MHz at room temperature.
- [c10] 10. The curable composition of claim 1, wherein said thermoplastic resin has been directly isolated from solution after polymerization.
- [c11] 11. The curable composition of claim 1, wherein said thermoplastic resin is a poly(phenylene ether).
- [c12] 12. The curable composition of claim 11, wherein said poly(phenylene ether) has a weight average molecular weight ranging from about 3,000 to 35,000 g/mol.
- [c13] 13. The curable composition of claim 11, wherein said poly(phenylene ether) has a weight average molecular weight ranging from about 3,000 to 35,000 g/mol.
- [c14] 14. The curable composition of claim 11, wherein said poly(phenylene ether) has been melt processed at a temperature ranging from about 200 ° to 350 ° C.
- [c15] 15. The curable composition of claim 11, wherein said poly(phenylene ether) is hydroxy functional.
- [c16] 16. The curable composition of claim 1, wherein said thermoplastic resin is one or more of a poly(phenylene ether) or a poly(styrene- co -maleic anhydride).
- [c17] 17. The curable composition of claim 1, wherein said thermoplastic resin is a reaction product of a poly(phenylene ether) and a peroxide.
- [c18] 18. The curable composition of claim 1, wherein said thermoplastic resin is a reaction product of a poly(phenylene ether), a peroxide, and a bisphenol.
- [c19] 19. The curable composition of claim 1, wherein said thermoplastic resin is a polyimide.
- [c20] 20. The curable composition of claim 1, wherein the curable composition further comprises one or more of an organic reinforcement, an inorganic

reinforcement, or a filler.

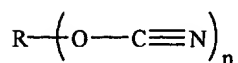
[c21] 21. The curable composition of claim 1, wherein the curable composition is essentially free of homopolymers of styrene.

[c22] 22. The curable composition of claim 1, wherein the epoxy resin is a multifunctional glycidyl ether.

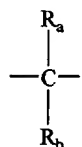
[c23] 23. The curable composition of claim 22, wherein said multifunctional glycidyl ether is selected from the group consisting of epoxidized phenol-formaldehyde novolacs, epoxidized cresol-formaldehyde novolacs, epoxidized alkylphenol-formaldehyde novolacs, epoxidized 1,1,1-tris(4-hydroxyphenyl)ethane, epoxidized 1,1,2,2-tetra(4-hydroxyphenyl) ethane, epoxidized phenol-dicyclopentadiene novolacs, and epoxidized phenol-benzaldehyde novolacs.

[c24] 24. The curable composition of claim 1, wherein the cyanate ester is selected from the group consisting of 1,3-dicyanatobenzene, 1,4-dicyanatobenzene, 2-tert-butyl-1,4-dicyanatobenzene, 2,4-dimethyl-1,3-dicyanatobenzene, 2,5-di-tert-butyl-1,4-dicyanatobenzene, tetramethyl-1,4-dicyanatobenzene, 4-chloro-1,3-dicyanatobenzene, 1,3,5-tricyanatobenzene, 2,2'-dicyanatobiphenyl, 4,4'-dicyanatobiphenyl, 3,3',5,5'-tetramethyl-4,4'-dicyanatobiphenyl, 1,3-dicyanatonaphthalene, 1,4-dicyanatonaphthalene, 1,5-dicyanatonaphthalene, 1,6-dicyanatonaphthalene, 1,8-dicyanatonaphthalene, 2,6-dicyanatonaphthalene, 2,7-dicyanatonaphthalene, 1,3,6-tricyanatonaphthalene, bis(4-cyanatophenyl)methane, bis(3-chloro-4-cyanatophenyl)methane, bis(3,5-dimethyl-4-cyanatophenyl)methane, 1,1-bis(4-cyanatophenyl)ethane, 2,2-bis(4-cyanatophenyl)propane, 2,2-bis(3,3-dibromo-4-cyanatophenyl)propane, 2,2-bis(4-cyanatophenyl)-1,1,1,3,3,3-hexafluoropropane, bis(4-cyanatophenyl)ester, bis(4-cyanatophenoxy)benzene, bis(4-cyanatophenyl)ketone, bis(4-cyanatophenyl)thioether, bis(4-cyanatophenyl)sulfone, tris(4-cyanatophenyl)phosphate, and tris(4-cyanatophenyl)phosphate.

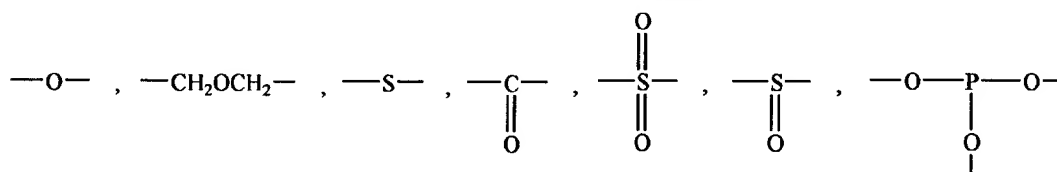
[c25] 25. The curable composition of claim 1, wherein the cyanate ester has the formula



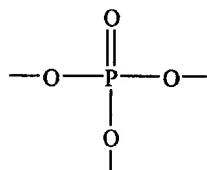
wherein R is an aromatic nucleus-containing residue which is selected from the group consisting of a residue derived from an aromatic hydrocarbon selected from the group consisting of benzene, biphenyl and naphthalene, a residue derived from a compound in which at least two benzene rings are bonded to each other by a bridging member selected from the group consisting of



wherein R_a and R_b are the same or different and each represents a hydrogen atom or an alkyl group containing 1 to 4 carbon atoms,



and

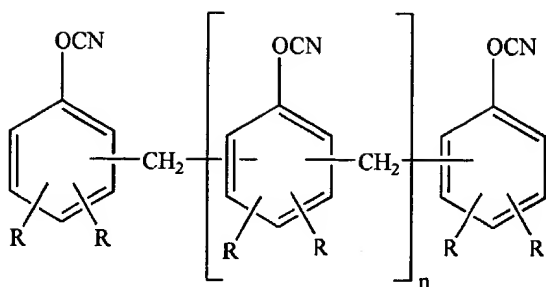


and a residue resulting from the removal of a phenolic hydroxyl group from a novolac-type or resol-type phenolic resin skeleton; said aromatic nucleus is optionally substituted by a substituent selected from the group consisting of alkyl groups containing 1 to 4 carbon atoms, alkoxy groups containing 1 to 4 carbon atoms, chlorine and bromine; n is an integer of 2 to 5; and the cyanate group is always directly bonded to the aromatic nucleus.

[c26] 26. The curable composition of claim 1, wherein the cyanate ester is a prepolymer of the cyanates esters of Claim 25, having a number average molecular weight of 400 to 6,000, and are formed by trimerizing the cyanate group of the cyanate esters.

[c27]

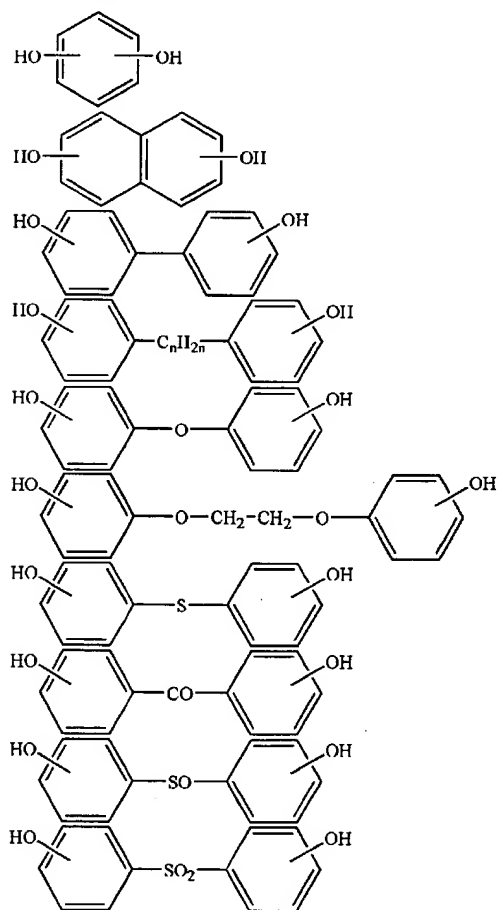
27. The curable composition of claim 1, wherein the cyanate ester is a cyanate-group-containing phenol resin comprising a mixture of polymers represented by the formula



wherein n is 0 or an integer of 1 or more; and R 's may be the same or different, and each R is a hydrogen atom or a methyl group, and containing 50% by weight or more in total of polymers having formula in which n is an integer of 1 to 3, the number average molecular weight of said phenol resin being 350 to 700 g/mole.

[c28]

28. The curable composition of claim 1, wherein the cyanate ester is a cyanic acid ester of an aromatic polycarbonate obtained by reacting an aromatic polycarbonate having one or two terminal hydroxyl groups with a cyanogen halide, wherein the aromatic polycarbonate is prepared from an aromatic dihydroxy compound represented by one of the following formulas:



where n is an integer of 1-4, inclusive, or a mono-, di-, tri- or tetra- halogeno-

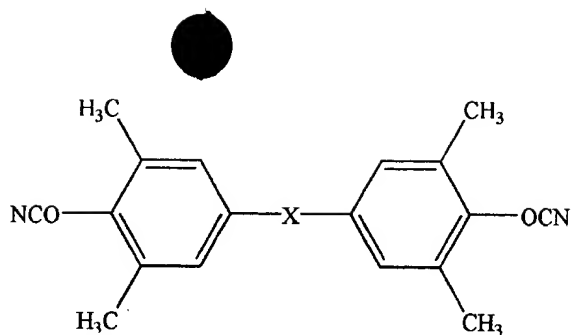
[illegible]

[c29]

where R is a divalent radical having 3 to 15 aromatic nuclei linearly linked together with ethereal oxygen atoms, said nuclei comprising nuclei selected from the group consisting of



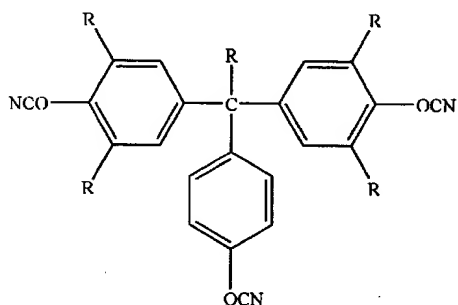
30. The curable composition of claim 1, wherein the cyanate ester has the structure



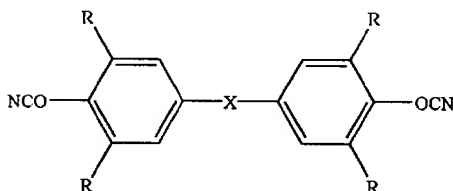
wherein X is methylene, isopropylidene, oxygen or divalent sulfur.

[c31]

31. The curable composition of claim 1, wherein the cyanate ester is a blend of a tricyanate ester and a dicyanate ester, wherein the tricyanate ester has the structural formula:



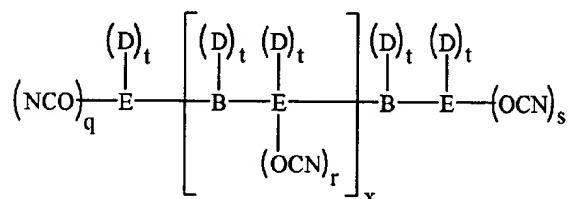
and the dicyanate ester has the structural formula:



wherein each R is H or methyl and is the same or different and wherein X is methylene, alkylidene having 2 to 4 carbon atoms, divalent oxygen, or divalent sulfur.

[c32]

32. The curable composition of claim 1, wherein the cyanate ester is a polyaromatic cyanate having the formula



wherein: E is an aromatic radical;

B is a C₇₋₂₀ polycyclic aliphatic radical;

D is independently in each occurrence any nonactive hydrogen-containing substituent;

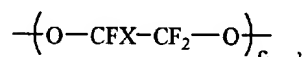
q, r and s are independently in each occurrence the integers 0, 1, 2, or 3; with

the proviso that the sum of q, r and s is greater than or equal to 2;
 t is independently in each occurrence an integer of between about 0 and 4
 inclusive; and
 x is a number between about 0 and 5 inclusive.

- [c33] 33. The curable composition of claim 1, wherein the cyanate ester is a
 fluorocarbon monocyanate having the structure

$$\text{F}_3\text{C}(\text{CFX})_a\text{A}(\text{CFX})_b\text{CH}_2\text{OCN}$$

where A is



X is fluorine or perfluoroalkyl having 1 to 10 carbon atoms, a is 1 to 10, b is 1,
 and c is 1 to 100.

- [c34] 34. The curable composition of claim 1, wherein the cyanate ester is a
 fluorocarbon dicyanate having the structure

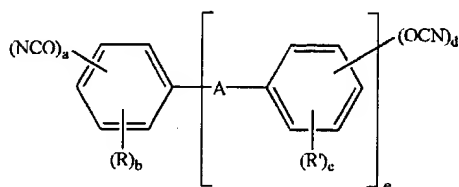
$$\text{NCOCH}_2(\text{CFX})_a\text{B}(\text{CFX})_b\text{CH}_2\text{OCN}$$

where B is (I) a carbon-to-carbon bond, in which case a is an integer of 1 to 30
 and b is zero, or (II) B is $[(\text{CFX})_d\text{O}(\text{CFX})_e]_f$, in which case a and b are zero, d
 and e are integers of 1 to 30, and f is an integer of 1 to 20, or (III) B is
 $(\text{OCF}_2\text{-CFX})_g\text{O}(\text{CFX})_h\text{O}(\text{CFX-CF}_2\text{O})_i$

in which case a and b are 1, h is an integer of 1 to 10, and g and i are integers
 of 1 to 100, or (IV) B is
 $[(\text{CF}_2\text{CH}_2)_j(\text{CF}_2\text{-CFX})_k]_m$,

in which case a and b are integers of 1 to 10, j and k are integers whose ratio
 j/k is 1/1 to 10/1, m is an integer of 1 to 100, and (CF_2CH_2) and $(\text{CF}_2\text{-CFX})$
 are randomly distributed units; and where X in all instances where it appears is
 fluorine or perfluoroalkyl of 1 to 10 carbon atoms.

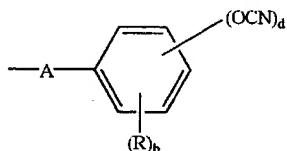
- [c35] 35. The curable composition of claim 1, wherein the cyanate ester has the
 formula



in which R represents hydrogen, halogen, linear or branched $\text{C}_1\text{-C}_9$ -alkyl or
 phenyl, two adjacent radicals R on the same nucleus together forming a

carbocyclic 5-membered or 6-membered ring or together and in conjunction with a hetero atom (O, S, N) forming a 5-membered or 6-membered heterocyclic ring, alkoxy radicals with 1 to 4 carbon atoms, alkoxy carbonyl radicals with 1 to 4 carbon atoms in the alkyl group;

R' has the same meaning as R or represents the group



where A is direct bond, a C₁-C₉-alkylene group optionally substituted by C₁-C₄-alkyl or phenyl, a cycloaliphatic or aromatic 5-membered or 6-membered ring, or a cycloaliphatic or aromatic 5-membered or 6-membered ring; a is a number from 0 to 5 where e = 1 and a number from 2 to 5 where e = 0; b = 5 - a where e = 1 and 6 - (a + d) where e = 0; c = 5 - d; d is a number from 0 to 5; e is the number 0, 1, 2 or 3; with the proviso that the sum of a and d (a + d) always gives a number from 2 to 5.

[c36]

36.A curable composition, comprising:

- (a) an epoxy resin and curing agent therefor, wherein said epoxy resin is a glycidyl ether resin or mixture of glycidyl ether resins containing, on average, greater than 2 epoxy groups per molecule;
- (b) 1,3,5-tris(2,4,6-tribromophenoxy)triazine and/or 2,2'-[(1-methylethylidene)bis[(2,6-dibromo-4,1-phenylene)oxy]]bis[4,6-bis[(2,4,6-tribromophenyl)oxy]-1,3,5-triazine];
- (c) a poly(phenylene ether) resin; and
- (d) a cyanate ester.

[c37]

37.A curable composition, comprising:

- (a) an epoxidized cresol-formaldehyde novolac resin;
- (b) 1,3,5-tris(2,4,6-tribromophenoxy)triazine;
- (c) a poly(phenylene ether) resin having a number average molecular weight ranging from about 1,000 to 15,000 g/mol; and
- (d) a cyanate ester.

[c38]

38.A cured composition comprising a cured residue of a curable composition comprising:

- (a) an epoxy resin and curing agent therefor, wherein said epoxy resin is essentially free of bromine atoms;
- (b) a flame retardant additive essentially free of phenolic groups and of epoxy groups, wherein said flame retardant is a condensation product of (i) a brominated phenol or a mixture of brominated phenols with (ii) a cyanuric halide;
- (c) a thermoplastic resin; and
- (d) a cyanate ester.

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